

The manuscript by Im et al. reports on the development and application of the UNIPAR model for prediction of SOA produced in smog chamber experiments by oxidation of toluene and 1,3,5-trimethylbenzene. While advancements in SOA modeling clearly are needed, particularly in the representation of particle-phase and heterogeneous reactions, in its current state the manuscript does not stand to advance the field to a great degree. The model likely is too computationally expensive and is highly empirical, making its application in regional/global chemical transport models unlikely and potentially irrelevant. Possibly a better goal of the model would be to provide insight into the relative importance of particle-phase and heterogeneous reactions (in this case, generalized oligomer formation and (inorganic) acid-catalyzed organosulfate formation) for SOA formation under a variety of atmospherically-relevant conditions (based on new or published chamber studies). With a significant amount of the relevant material in the supplemental section, the manuscript is somewhat hard to follow. And again, the relevant conclusions and implications are lacking. It is recommended that the manuscript be reevaluated after significant revision.

#### Scientific Comments:

Abstract-The authors mention the use of the SOA partitioning model in CMAQv5.0.1 (newest release); however, it is not clear what (if any) unique features of this model are being applied. The SOA model in CMAQv.5.0.1 has its own treatment of oligomer formation and non-volatile SOA formation via oxidation of aromatic compounds under low-NO<sub>x</sub> conditions. Comparison with the CMAQ SOA model may be one way in which the manuscript could be improved. It rather appears that standard gas/particle partitioning theory is being applied.

p. 5845 line 10-The VBS model does not account for oxidation reactions in the condensed phase.

p. 5850 line 21-The model assumes ideality for compounds condensing into the organic phase, while estimating partitioning coefficients for compounds partitioning into the inorganic/aqueous phase, citing Zuend and Seinfeld, 2012. However, Zuend and Seinfeld 2012 indicate that treating particles as two separate phases at high RH (which applies to the chamber experiments reported) is not justified—rather the particles likely exist as a single phase. In addition, the O:C ratio is an important parameter for understanding mixing and phase separation (in the absence of detailed modeling). The manuscript does not address predicted/measured changes in O:C ratios (say as a function of oligomerization), which may have an impact on the assumed two-phase system, as well as the activity coefficients of condensing compounds (i.e., is the nature of the condensed phase very different if dominated by monomers vs. dimers and how is the partitioning of each affected). Zuend and Seinfeld conclude that assuming ideality at high RH (>60%) greatly overpredicts SOA mass (if the system has relatively low O:C ratios). It is recommended that the authors consider O:C ratios and the effects on partitioning/phase separation, and potentially the prediction of activity coefficients for partitioning into an organic phase (if not justified to assume phase separation and/or ideality in the organic phase).

S6-If I understand correctly, the possible list of products is the same (albeit differently weighted-different alpha values) under low vs. high NO<sub>x</sub> conditions. Is this consistent with chamber studies in which products have been measured? Is the initial product distribution determined from a low-NO<sub>x</sub>

MCM run and then fitting done with decreasing VOC/NO<sub>x</sub> ratio? Assuming that is the case, what happens if initial predictions are done for high-NO<sub>x</sub> conditions and then fitting with increasing VOC/NO<sub>x</sub> ratio?

Figure S9-The model simulations show that for toluene, the oligomer fraction decreases as NO<sub>x</sub> decreases. However, Ng et al. (2007) report that under low-NO<sub>x</sub> conditions, the SOA that forms is effectively non-volatile (suggesting a greater extent of oligomer formation). This is not discussed anywhere in the manuscript.

Editorial Comments:

p. 5846 line 10-Need reference(s) for CMAQ model.

p. 5850 line 14-Not sure why  $OM_T$  is used instead of  $M_{or}$  (since  $M_{in}$  is used for total inorganic mass).

p. 5852 line 20-The authors state that if  $RH < ERH$ , the particle is crystalized and acid-catalyzed reactions can't occur. However, they go on to state that the inorganic phase reaction was restricted to the dry condition ( $RH < ERH$ ).

It is suggested that the authors reconsider the order of the supplement material (e.g., section S5 should go before S4).